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(21) International Application Number: PCT/US98/16489 (22) International Filing Date: 7 August 1998 (07.08.98) (30) Priority Data: 60/055,056 8 August 1997 (08.08.97) US (71) Applicant: E.I. DU PONT DE NEMOURS AND COMPANY [US/US]; 1007 Market Street, Wilmington, DE 19898 (US). (72) Inventor: POWELL, Richard, James; 4217 Meeks Drive, Orange, TX 77632 (US). (74) Agent: KELLY, Patricia, L.; E.I. du Pont de Nemours and Compagny, Legal Patent Records Center, 1007 Market Street, Wilmington, DE 19898 (US).		(81) Designated States: CA, JP, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
(54) Title: DELAMINATION PEEL SEAL WITH IMPROVED ANTIBLOCK PROPERTIES		
(57) Abstract This invention provides delamination peel seal layers and structures that demonstrate a reduced tendency to block. The delamination peel seal layers and structures are formed from ethylene/acid copolymers and/or ionomers, with the addition of high density polyethylene to reduce blocking. Such layers and structures are useful in packaging applications, such as cereal box liners, that require a peelable seal.		

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TITLEDELAMINATION PEEL SEAL WITH IMPROVED
ANTIBLOCK PROPERTIES

5 This application claims the benefit of U.S.
Provisional Application No. 60/055,056, filed
August 8, 1997.

FIELD OF THE INVENTION

10 This invention relates to delamination peel
seal layers and structures comprising an ethylene/acid
copolymer or an ionomer. This invention specifically
relates to delamination peel seal layers and structures
further comprising high density polyethylene which
demonstrate reduced blocking. Such layers and
15 structures are useful in packaging applications that
require a peelable seal.

BACKGROUND OF THE INVENTION

 Ethylene/acid copolymer and ionomer films and
resin coatings can exhibit a tendency to block during
20 processing of these materials. Whether processed as a
film itself, as a film layer coextruded with other
layers, or as a coating on a film, surfaces of these
materials will often stick together during blown film
production. This makes separation of the layers for
25 further use difficult.

 Film blocking forces can cause tearing or poor
mill roll quality during the in-line slitting,
separation and wind-up process in blown film
production. In these cases the line must be slowed to
30 reduce the stresses, but this in turn limits
productivity. High blocking forces also retard or even
prevent slip additive migration to the surface or
interface and this can result in poor performance in
smoothly moving through packaging or bag forming
35 equipment.

 A common way to reducing blocking is by the
addition of anti-block additives. Silicas, such as
diatomaceous earth or silica dioxide particles are

commonly used as antiblock additives for polyethylene, ethylene/(meth)acrylic acid copolymers and ionomer films and coatings. Levels used are of the order of 0.3 to 1.5% by weight, usually added by incorporating concentrates containing 10 to 20% silica. These concentrates are expensive and add to the expense of the final product. In addition, silica dioxide particles are very abrasive and can add to the cost of maintaining sharp knives in film slitting, as well as in underwater pellet cutters.

There is a need for a lower-cost alternative to conventional antiblock additives for ethylene/acid copolymer and ionomer films and coating resins used in delamination peel seal layers and structures.

SUMMARY OF THE INVENTION

The invention herein provides a delamination peel seal layer comprising:

- (a) an acid copolymer or an ionomer, or blend thereof, wherein the acid level of the acid copolymer ranges from about 8% to about 15% by weight, and the ionomer is up to about 80% neutralized, and
- (b) about 1% to about 15% by weight of high density polyethylene.

The invention also provides a delamination peel seal structure comprising a seal layer coextruded with a thermoplastic film, wherein the seal layer comprises

- (a) an acid copolymer or an ionomer, or blend thereof, wherein the acid level of the acid copolymer ranges from about 8% to about 15% by weight, and the ionomer is up to about 80% neutralized, and
- (b) about 1% to about 15% by weight of high density polyethylene.

Optionally the peel seal layer may also comprise a slip additive or other useful additives, such as antioxidants.

DETAILED DESCRIPTION

Ethylene/acid copolymers and ionomer films and resin coatings have been found to be useful in delamination peel seal layers and structures. A

5 "delamination peel seal layer" may be defined as an adhesive layer that is heat-sealable, used to firmly adhere two structures or layers together, though remain peelable at a designed strength. Though not limited to any one peel mechanism, the delamination peel seal
10 layer typically peels by breakage or tearing through the peel seal layer, followed by controlled strength peeling from a high density polyethylene or other thermoplastic substrate layer.

A "delamination peel seal structure" comprises
15 a delamination peel seal layer in combination with substrates, structures or layers that are to be sealed or joined. For example, a delamination peel seal structure may include a delamination peel seal layer coextruded with a thermoplastic film, e.g. a layer of
20 high density polyethylene (HDPE). The HDPE layer may form a package or container that is sealed at an opening by a delamination peel seal layer.

Ethylene/acid copolymers, or simply acid copolymers, and their corresponding ionomers are well
25 known in the art to be copolymers of ethylene with an olefinically unsaturated organic mono- or di-acid such as acrylic or methacrylic acid, or maleic acid or fumaric acid or their anhydrides, the acid (or anhydride) typically comprising about 0.5 to 50 mole
30 percent of the total polymeric material. Ethylene/acid copolymers and their methods of preparation are well known in the art and are disclosed in, for example, U.S. Pat. Numbers 3,264,272, 3,404,134, 3,355,319 and 4,321,337, incorporated by reference herein.

35 Ethylene/acid copolymers useful in the delamination peel seal layer and structure of the invention herein, typically have an acid content

ranging from about 8% to about 15% by weight,
preferably about 9% to about 14% by weight.

Ethylene/acid/acrylate terpolymer and
corresponding ionomers are well known in the art to be
5 copolymers of ethylene, an olefinically unsaturated
organic acid such as acrylic or methacrylic acid and an
alkyl acrylate or methacrylate monomer (e.g. n-butyl
acrylate or methacrylate or isobutylacrylate).

The delamination peel seal layer or structure of
10 the invention herein may comprise acid copolymers,
corresponding ionomers, or blends thereof. In this
disclosure, the word "copolymer" means a polymer
polymerized from two or more monomers, and includes
terpolymers. The more specific description, e.g.
15 'ethylene methacrylic acid copolymer', and the like, is
meant to include copolymers which may also have a third
monomer present.

The acid copolymers are termed ionomers when the
acid is neutralized in whole or in part to produce a
20 salt. The cations for said salts are usually an alkali
metal such as sodium, potassium, or the like, or
divalent or trivalent metal ions. Suitable acid
copolymers and ionomers are available from the DuPont
Company, Wilmington, DE, under the trade names Nucrel®
25 and Surlyn®, respectively.

There are three key compositional features to the
ionomers suitable for this invention. They are the
average acid level of the ethylene-acid copolymers, the
average level of neutralization for corresponding
30 ionomers, and the type and relative amounts of metal
ions with which the materials are neutralized.

The average weight percent acid of the
corresponding or underlying acid copolymers should be
between about 8% and about 15%, preferably between
35 about 9% and 14%. It will be recognized that a blend
with this level of acid can be obtained by blending
copolymers, one or both of which has an acid level

outside this range. Preferably the weight percent acid in each acid copolymer, and those from which the ionomers are derived should be close to the 8% to 15% range, and most preferably they should be within this range.

The average level of neutralization of the acid groups in the underlying ethylene/acid copolymers will range up to about 80%, and should be greater than about 10 percent, though preferably not higher than about 35 percent. More preferably, the average level of neutralization is from about 20% to about 30%.

As mentioned above, a delamination peel seal layer or structure may comprise blends of ionomers or ionomers and acid copolymers. Commercially produced ionomers may have their underlying acid groups neutralized to between about 10 and 90 percent. The conventional definition of ionomers, as defined in U.S. Pat. No. 3,264,272 (Rees) states that an ionomer is an acid copolymer whose acid groups are from 10 to 90% neutralized. It will be recognized that an individual ionomer used to make a blend may be less than 10% neutralized, but that on blending with an ionomer of higher level of neutralization, the desired average level of neutralization can be achieved. It would even be possible to incorporate additional unneutralized acid copolymer and be within the bounds of the definition of the invention. In the melt, the ions in an ionomer are believed to be quite labile. As a result, the metal ions become distributed and associated with the acid groups of the polymer chains present. Thus, a polymer chain derived from an ionomer with a low level of neutralization will not remain at that low level of neutralization if the average level of neutralization is higher.

The third compositional requirement concerns the particular metal ions and their relative amounts in the blend. Suitable metal ions used to neutralize the underlying ethylene/acid copolymers include sodium,

potassium, zinc, magnesium and calcium. Mixed ion systems are also possible.

Since full reaction of the acid and ion occurs, the amount of metal ion present defines the degree of neutralization, discussed above. The effective neutralization is adjusted between 0 and 80% by varying the amount of metal ions and acid level to control adhesion to a substrate at a desired level. Generally lower acid levels will require higher neutralization than higher acid levels when controlling adhesion to a high density polyethylene substrate in a peel seal structure.

Because the ions are labile in the melt, a composition containing a specific degree of neutralization and a mixture of metal ions may be obtained in several ways. The simplest way is by blending different metal ionomers each containing sufficient of each of the metal ions to provide the desired ratio of ion types. However, ionomers which contain more than one type of ion or ionomers with low amounts of a given ion, together with one with a high amount of that ion, and even an un-neutralized acid copolymer, can be blended to make a given blend, provided the final degree of neutralization is satisfied.

The delamination peel seal layer of the invention herein further comprises from about 1% to about 15% by weight high density polyethylene (HDPE). The HDPE is added to reduce the blocking tendency of the acid copolymer or corresponding ionomer. The HDPE is preferably present in an amount ranging from about 2% to about 10% by weight. The density of the HDPE preferably ranges from about 0.941 to about 0.960 g/cc and higher, as defined by Types III and IV in ASTM D-1248. More preferably, the density ranges from about 0.945 g/cc to about 0.965 g/cc. The melt index of the HDPE preferably ranges from about 0.1 to about 500, more preferably from about 0.5 to about 20. The melt

index is typically measured following the procedure of ASTM D-1238.

Optionally, the delamination peel seal layer may comprise additional components, such as slip additives. Suitable slip additives include n-oleyl palmitamide, stearamide, benhenamide, and the like. Other useful additives, such as antioxidants, are also contemplated.

The invention herein also provides a delamination peel seal structure. The structure comprises a seal layer (as described above) coextruded with a thermoplastic film substrate. Suitable thermoplastic films include films of nylon, polypropylene and polyester and preferably high density polyethylene. In materials where the delamination bonding strength cannot directly be obtained between the peel seal layer and substrate, as disclosed herein, then a third coextrudable adhesive layer between the thermoplastic and acid copolymer or ionomer can be used to control the bond strength to the desired level.

The delamination peel seal layer and structure of this invention are particularly useful in packaging applications where a heat-sealable, yet peelable, seal is desired. Such applications include cereal box liners, cracker and cookie packaging, or any dry food package requiring a peelable seal. Typically, a cereal box liner is made by coextruding an acid copolymer or ionomer layer with a layer of HDPE and forming the resulting laminate into a package. The acid copolymer or ionomer surfaces of the resulting laminate can be sealed together at the package opening by the application of heat and pressure. The resulting seal is peelable. The strength of the peel seal may be varied by varying the composition of the acid copolymer or ionomer, by coextrusion conditions and/or by the seal layer thickness. The seal layer thickness typically varies between about 4 and 50 micrometers.

The invention herein overcomes the common problem of blocking encountered in the production of such packages. The addition of HDPE to the acid copolymer or ionomer reduces blocking to a significant extent.

- 5 The HDPE may be added to the acid copolymer or ionomer resin itself by melt blending into the acid copolymer or ionomer prior to pellet production or into the sealant film forming extruder feed, along with the resin, as a separate pellet source or polymer feed.

- 10 The invention will be further clarified by the following examples, which are intended to be purely exemplary of the invention.

EXAMPLES

The following test methods were used for the examples:

- 15 Melt Index ASTM D-1238 Condition 190/2.16
Gloss, 20° ASTM D-2457
Haze, % ASTM D-1003
Coefficient of
Friction (COF) ASTM D-1894

- 20 Block, in gm/sq. in, was determined by the following method. The blocking force in shear was measured by pulling apart in a parallel planar direction the layers of the blown film bubble which have been pressed together by going through the nip roll. A blocked
25 sample area of four square inches (25.8 cm) is measured. The absolute values will vary with the specific blown film equipment used as well as the temperature at the nip roll.

COMPARATIVE EXAMPLE A and EXAMPLES 1-3

- 30 A series of blends were made by adding high density polyethylene (HDPE) of 0.45 melt index (MI) and 0.950 g/cc density, containing a small amount of octene comonomer, to a sodium ionomer of ethylene/methacrylic acid to determine an effective level to improve or
35 prevent film blocking.

Comparative Example A contained 0.7% silica by weight. This was added from a 20% silica concentrate in an ethylene/methacrylic acid copolymer (MAA) of 12 MI

and 8.7% MAA. This blend was melt compounded in a 2 inch (5.08 cm) diameter single screw extruder at about 250°C melt temperature. The silica in the concentrate was 6 micron "Superfloss" diatomaceous earth from Celite Company.

The blends containing HDPE did not contain any silica. The ionomer used was a nominal 1.8 melt index containing 9.5 wt% MAA and neutralized 27% with sodium ion. This ionomer was obtained by modification of a more highly neutralized ionomer (about 53%) during the melt blending by adding an acid copolymer to effectively reduce its degree of neutralization to provide the final blend composition desired. This gives the identical properties as though the lower neutralized ionomer specified above was separately prepared.

Each of the melt blends were in turn dry blended at room temperature with 8.5% pellets of a slip additive concentrate. The slip additive was stearamide and was added via a concentrate of 20% stearamide in a 10 melt index, 8.7% MAA ethylene/methacrylic acid copolymer. These dry blends were then blown into 2.0 mil film from a 1.5 inch (3.8 cm) diameter extruder at 230°C melt temperature and 1.9:1 blow up ratio.

The data in Table I shows that equivalent antiblock protection is achieved at about 2-3% HDPE compared to 0.7% silica. No adverse effect on heat seal strength or hot tack was observed.

TABLE I

Example	HDPE %	Gloss 20°	Haze %	COF	BLOCK g/sq.in.
A	0	74	3.8	0.2	>114
1	2	94	4.2	> 1	>114
2	4	77	7.7	0.15	17
3	6	50	13.8	< 0.1	0

NOTE: 114 g/in² = 17.67 g/cm²; 17 g/in² = 2.64 g/cm².

COMPARATIVE EXAMPLE B and EXAMPLES 4-5

For Example 4, a melt blend of sodium ionomer of ethylene/methacrylic acid copolymer and 6% of high density polyethylene (HDPE) were made in a single screw extruder at about 250°C melt temperature. The HDPE and the ionomer were equivalent to that used in Example 1.

For Example 5, a melt blend similar to Example 4 was made only the 6% HDPE was from a different supplier and contained no comonomer. The melt index of the HDPE was 0.46 and density was 0.960 g/cc.

In Comparative Example B, a similar formula was made but "Superfloss" silica was added at the 0.7% level in place of the HDPE. This was added from a 20% silica concentrate in an ethylene/methacrylic acid copolymer (MAA) of 12 MI and 8.7% MAA. This was also homogenized by melt blending in a single screw extruder at 250°C melt temperature.

Each of the above blends were in turn dry blended at room temperature with 8.5% pellets of a slip additive concentrate. The slip additive was stearamide and was added via a concentrate of 20% stearamide in a 10 Melt Index, 8.7% MAA ethylene/methacrylic acid copolymer. These dry blends were then blown into 2.0 mil film from a 1.5 inch (3.8 cm) diameter extruder with a 1.9:1 blow up ratio.

TABLE II

Example	MELT INDEX	Gloss 20°	Haze %	COF	BLOCK g/sq.in.
B	2.2	84	3.6	0.21	255
4	2.0	42	16.5	0.13	0
5	2.0	46	14.9	0.12	0

NOTE: 255 g/in² = 39.53 g/cm².

Other embodiments of the invention will be apparent to those skilled in the art from consideration of the specification and practice of the invention disclosed herein. It is intended that the specification and examples be considered as exemplary

only, with a true scope and spirit of the invention
being indicated by the following claims.

WHAT IS CLAIMED IS:

1. A delamination peel seal layer comprising:
 - (a) an acid copolymer or an ionomer, or blend thereof, wherein the acid level of the acid copolymer ranges from about 8% to about 15% by weight, and the ionomer is up to about 80% neutralized, and
 - (b) about 1% to about 15% by weight of high density polyethylene.
2. The delamination peel seal layer of claim 1 further comprising a slip additive.
3. The delamination peel seal layer of claim 1 wherein the acid level of the acid copolymer ranges from about 9% to about 14% by weight, and the ionomer is about 10% to about 35% neutralized.
4. The delamination peel seal layer of claim 1 wherein the acid level of the acid copolymer ranges from about 9% to about 10% by weight, and the ionomer is about 20% to about 30% neutralized.
5. The delamination peel seal layer of claim 1 wherein the high density polyethylene comprises about 2% to about 10% by weight.
6. The delamination peel seal layer of claim 1 wherein the high density polyethylene has a density of at least about 0.941 g/cc.
7. The delamination peel seal layer of claim 6 wherein the high density polyethylene has a density ranging from about 0.941 to about 0.965 g/cc.
8. The delamination peel seal layer of claim 1 wherein the high density polyethylene has a melt index ranging from about 0.1 to about 500.
9. The delamination peel seal layer of claim 8 wherein the high density polyethylene has a melt index ranging from about 0.5 to about 20.
10. The delamination peel seal layer of claim 1, wherein the ionomer is an ethylene/methacrylic acid copolymer, having about 9.5 weight percent methacrylic acid and being about 27% neutralized, and the high density polyethylene exhibits a melt

index of about 0.45, a density of about 0.950 g/cc and comprises about 4% to about 8% by weight of the seal layer.

- 5 11. A delamination peel seal structure comprising a seal layer coextruded with a thermoplastic film, wherein the seal layer comprises
- 10 (a) an acid copolymer or an ionomer, or blend thereof, wherein the acid level of the acid copolymer ranges from about 8% to about 15% by weight, and the ionomer is up to about 80% neutralized, and
- (b) about 1% to about 15% by weight of high density polyethylene.
- 15 12. The delamination peel seal structure of claim 11 wherein the acid level of the acid copolymer ranges from about 9% to about 14% by weight, and the ionomer is about 10% to about 35% neutralized.
- 20 13. The delamination peel seal structure of claim 11 wherein the high density polyethylene in the seal layer comprises about 2% to about 10% by weight.
14. The delamination peel seal structure of claim 11 wherein the high density polyethylene in the seal layer has a melt index ranging from about 0.1 to about 500.
- 25 15. The delamination peel seal structure of claim 11 wherein the high density polyethylene in the seal layer has a melt index ranging from about 0.5 to about 20.
- 30 16. The delamination peel seal structure of claim 11 wherein the ionomer is ethylene/methacrylic acid copolymer, having about 9.5 weight percent methacrylic acid and being about 27% neutralized, and the high density polyethylene exhibits a melt index of about 0.45, a density of about 0.950 g/cc and comprises about 4% to about 8% by weight of the seal layer.
- 35 17. The delamination peel seal structure of claim 11 wherein the thermoplastic film comprises high density polyethylene.

18. A package comprising the delamination peel seal structure of claim 11.
19. A cereal box liner comprising the delamination peel seal structure of claim 11.
- 5 20. A process for reducing the blocking tendency of a resin comprising an acid copolymer, an ionomer, or blend thereof, in a delamination peel seal layer, comprising the step of adding about 1% to about 15% by weight HDPE to the resin by melt blending or into a sealant film forming extruder as a separate polymer feed along with the resin, wherein the acid copolymer has an acid level ranging from about 8% to about 15% by weight, and the ionomer is up to about 80% neutralized.
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- 15

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 98/16489

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08L23/08 C09J123/08 B32B27/32

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

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IPC 6 C08L C09J B32B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 574 108 A (ANTONOV VLADIMIR ET AL) 12 November 1996 see column 3, line 64 see column 4, line 22 - line 24 see column 4, line 34 - line 36; claims 1-4; table 1	1-18, 20
X	DE 196 10 415 A (SCHULMAN A GMBH) 18 September 1997 see page 3, line 44 - line 46; claims 1, 2, 5, 14	1-17
Y	US 4 550 141 A (HOH GEORGE L) 29 October 1985 see column 14, line 13 - line 15; claims 1-10	1-20
	-/-	

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 98/16489

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	<p>US 5 160 767 A (GENSKE ROGER P ET AL) 3 November 1992 see column 2, line 23 - line 28 see column 3, line 46 - line 50 -----</p>	1-20

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 98/16489

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